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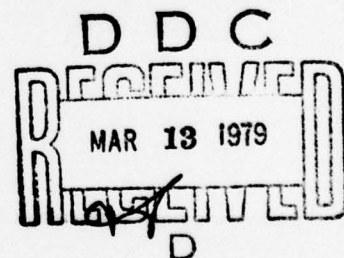
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2650 Å ULTRAVIOLET LASER BY HARMONIC GENERATION

By

Lu Shaw-Shin, Hsu Ta-Shun



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RESEARCH REPORT

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2650 A Ultraviolet Laser by Harmonic Generation*

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Ultraviolet laser has wide applications in the fields of photochemistry, biochemistry, high energy photon research and agriculture. Since the efficiency of self emission radiation is proportional to the third power of frequency, it is generally difficult to obtain ultraviolet laser through direct stimulation; and hence nonlinear optics crystals are often used in the generation of ultraviolet laser based on their frequency doubling effect.

April to July 1975, two members of the Optics Department of Shantung University, Lu Shaw-Shin and Hsu Ta-Shun, did their post graduate training with the Nonlinear Optics Group of the Laser Research Laboratory at the Physics Research Institute, China Academy of Sciences. During this period, we studied the generation of 2650 A^o ultraviolet laser by the second harmonic of the 1.06 μ infrared laser light from a neodymium glass laser. This article is a brief introduction to that work.

I. Experimental Arrangement

A rotating mirror Q - switched neodymium glass laser is used as the fundamental frequency light source where the working material is a ϕ 16 x 250 mm neodymium glass rod, angular speed of the rotating mirror

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is 10,000 rev/min after doubled angular acceleration, the transmissivity of the output mirror is 60 percent, the length of the resonance cavity is 500 mm. The focusing cavity is a dual spherical cavity with double pumping lamps, the measured output laser pulse width is 50 ns and its peak power is 40 MW.

The 1.06 μ laser enters the first frequency doubling crystal after passing through a glass plate set at the Brewster angle, the polarized \leftarrow component useful for frequency doubling suffers essentially no loss, while the small fraction of laser reflected by the glass plate enter a power meter which monitors the fundamental frequency output and its intensity fluctuation during the experiment.

The first frequency doubler crystal is LiIO_3 of the $C_6 - 6$ group and its transmissivity region, according to the Laser Handbook, is approximately 0.3 μ to 6 μ . LiIO_3 has the advantage of having relatively large nonlinear coefficients.⁽¹⁾ It has the following nonzero nonlinear coefficients:

$$d_{15} = d_{24} = d_{31} = -d_{32} \text{ and } d_{33},$$

$$d_{31} (\text{LiIO}_3) = (11.9 \pm 1.0) d_{34} (\text{KDP}).$$

LiIO_3 has phase matching of the first kind only ($o + o \rightarrow e$), and the calculated matching angle $\theta_M = 29.8^\circ$ is in agreement with the experimental result. The LiIO_3 crystal is 15 mm thick, cut according to θ_M , and coated with monolayer film which also provides damp proof effect. A filter is placed behind the LiIO_3 crystal to filter out the fundamental frequency light of 1.06 μ and let through the frequency doubled light (5300 Å). The 5300 Å laser is then focused with lenses and incidents on the second frequency doubler crystal.

We have chosen ADP as the second frequency doubler mainly because it has much higher transmissivity in the ultraviolet region than other nonlinear crystals. The Laser Handbook data showed that the transparent region of ADP is approximately $0.125 - 1.7 \mu$. ADP has the added advantage that the optimum (90°) phase match is easily realized for the doubled frequency 5300 \AA by keeping the ADP at a certain temperature. From the empirical equation (2)

$$n^2 = A + B \nu^2 / (1 - \nu^2/C) + D/(E - \nu^2),$$

where N is the refractive index, ν is the wave number, A , B , C , D , and E are empirical constants, we obtain the refractive index of ADP, shown in Table 1.

TABLE I Refractive Index of ADP

λ	n_o	n_e
5300 \AA	1.5277	
2650 \AA	1.5797	1.5266

Other useful empirical equations are (3):

$$n(T) = n(298K) + \Delta n,$$

$$\Delta n = (n^2 + an + b) c \cdot \Delta T,$$

where $\Delta T = (298 - T)K$ and a , b , c are empirical constants; for ADP, their values are shown in Table 2.

90° phase matching is achieved at the phase matching temperature

T_{PM}

$$n_o(5300 \text{ \AA}) = n_e(2650 \text{ \AA}),$$

and T_{pM} is calculated to be

$$T_{pM} = 325.4 \text{ K} = 52.4^{\circ} \text{ C.}$$

TABLE 2 Empirical constants of ADP

	a	b	$c(K^{-1})$
• 光	-3.0297	2.3004	0.713×10^{-4}
• 光	0	0	0.675×10^{-4}

In order to control the ADP temperature near T_{pM} , we have designed and built a constant temperature up to 100° C with an accuracy better than 0.1° C and a temperature gradient less than 0.1° C across the volume containing the ADP crystal. The heating rate should be kept less than 5° C/min to avoid excessive temperature shock to the crystal.⁽⁴⁾

ADP belongs to the 42m crystal group with nonzero nonlinear coefficients $d_{36} = d_{25} = d_{14}$, and for a 90° phase matching, the crystal should be 45° Z cut. We have kept the cross section of the crystal relatively small (8 mm x 8 mm) to ensure a uniform temperature across the cross sectional area, and the length of the crystal is 60 mm. Since the cross section of the light beam is greater than the cross sectional area of the crystal, the 5300 Å laser beam is focused which also increases the efficiency of frequency doubling. The output mode of the neodymium laser is very complicated, hence an accurate calculation of the optimum focal length of the lens is difficult. However, we can assume a Gaussian beam and estimate its value. According to the theory of G.D. Boyd and

D. A. Kleinman ⁽⁵⁾, the optimum focusing condition is $1/b = 2.84$, where l is the length of the crystal and $b = w_0^2 k$, the common focal parameter. The radius of the focused beam, w_0 , is therefore 3.42×10^{-3} cm. The half divergence angle of our light beam is 1-2 mrad, a converging lens of a few centimeters of focal length is required to focus the light beam to a spot of 3.42×10^{-3} cm in the crystal. This is not possible in practice mainly because the focal length has to be greater than the half length of the constant temperature oven; furthermore, the excessive power density of a small spot will cause damage to the crystal. After repeated tests, we have confirmed that a lens of 420 mm focal length does not lead to crystal damage and used such a lens in our experimental set up, shown in Fig. 1.

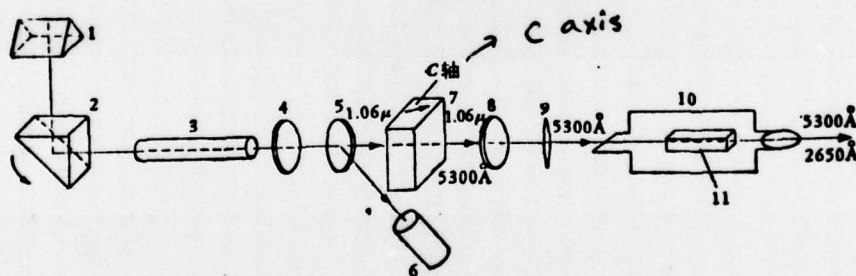


Fig. 1 Schematic diagram of the experimental set up. 1. Fixed total reflection prism, 2. rotating prism, 3. neodymium glass rod, 4. output mirror, 5. glass plate, 6. fundamental frequency monitor, 7. LiIO_3 crystal, 8. filter, 9. focusing lens ($f = 420$ mm), 10. constant temperature oven, 11. ADP crystal.

II. Results and Discussion

We have first placed a quartz prism spectrograph behind the oven and photographed the 5300 Å laser spectra at various ADP temperatures, as shown in Fig. 2. We have noticed that, with increasing crystal temperature, a new spectrum line appeared on the long wavelength side of the 2650 Å line, and, as the temperature is increased, the new line moves toward longer wavelength. This effect warrants further investigation and we studied the phenomena more closely. Our finding is as follows: when the crystal temperature is in the 45 - 46° C range, one strong spectrum line shows up; for crystal temperatures above or below this value, two lines were observed in most cases. One of the two lines is essentially fixed in position, coincide with the singlet line position at 45 - 46° C; the other line, however, shifts its position with changing temperature. When the temperature is above 45 - 46° C and rising, the

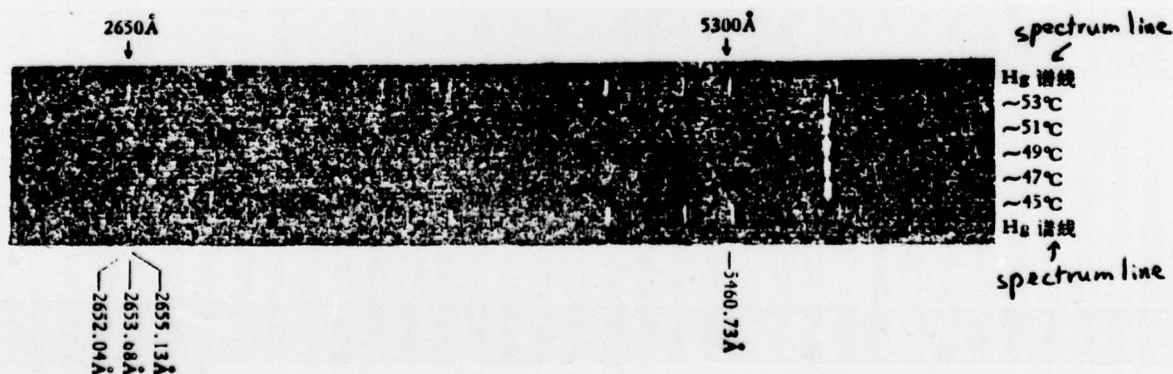


Fig. 2 Laser spectrum of 5300 Å and 2650 Å.

line shifts toward longer wavelength; for temperatures below $45 - 46^{\circ} \text{C}$ and dropping, the shift is toward shorter wavelength. The intensity of the moving spectrum line is greater than that of the fixed line. Occasionally, only the moving spectrum line is observed.

In order to explain this phenomenon, we have carried out more experimentation and our preliminary interpretation is that the shift is due to the finite spectrum distribution of the fundamental frequency laser light. More rigorous interpretation can be made by photographing the fine structures of the 1.06μ fundamental and the 5300 \AA doubled-frequency spectra. Circumstances did not allow further experiments along this line.

Subsequently, we resolved the 2650 \AA and the 5300 \AA laser light with the quartz prism in the spectrograph, and, with suitable filters attenuating the ultraviolet light, we measured the relative intensity of the ultraviolet laser light with photomultipliers. We found that the intensity of the output ultraviolet has several tens of percents fluctuation under the conditions of a constant fundamental frequency intensity. Our data are averaged and normalized (see Table 3) before they were plotted as the normalized relative intensity versus temperature curve, shown in Fig. 3. The optimum phase matching temperature is then obtained from Fig. 3, $T_{PM} \approx 45.4^{\circ} \text{C}$, and the half-maximum full-width of the relative intensity - temperature curve is $\Delta T_{1/2} \approx 4.7^{\circ} \text{C}$.

Discrepancy exists between the measured and computed values of T_{PM} . This is not surprising, however, as pointed out by B.G. Huth and Y.C. Kiang (6), there is systematic error in computations using the Phillips empirical equation and the computed T_{PM} is greater than the measured T_{PM} at temperatures above room temperature. In addition, our

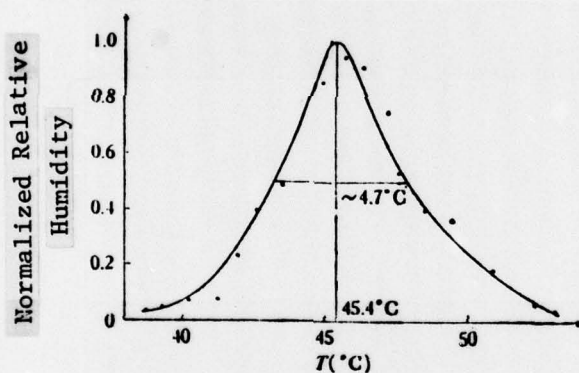
Table 3. Relative intensity - temperature relationship of ultraviolet laser light.

$T(^{\circ}\text{C})$	38.7	39.3	40.2	41.2	41.9	42.6	43.5	44.2	44.9	45.2	45.7	46.3	47.2	47.6	48.5	49.4	50.8	52.3	53.0
(1) 相对强度	1.7	2.4	3.9	4.2	13	22	27	40	47	55	52	50	41	29	22	20	9.6	3.2	1.7
(2) 归一化	0.031	0.044	0.071	0.076	0.236	0.400	0.491	0.727	0.855	1	0.945	0.909	0.745	0.527	0.400	0.364	0.181	0.058	0.031

(1) relative intensity

(2) normalized

Fig. 3 Ultraviolet laser normalized relative intensity as a function of temperature variation.



results are in agreement with results reported in foreign literatures.

Using Nd:YAG laser, D. P. Schinke measured the T_{pm}^{pm} of ADP used in the 5320 Å to 2660 Å frequency doubling to be 49.5°C (7). The center wavelength of our green laser is 5300 Å and, according to the curve given by Huth and Kiang (6), the phase-matching temperature drops approximately 30 degrees for every 100 Å decrease in the fundamental wavelength, therefore, our T_{pm}^{pm} should be 3 or 4 degrees below 49.5°C .

Our measured value is 4.1°C below 49.5°C , consistent with the Schinke result.

The measured $\Delta T_{\frac{1}{2}}$ in our experiments is a relatively wide 4.7°C . This is mostly due to the relatively large width of the neodymium glass laser and, as a result, there is always some frequency component satisfying the phase matching condition over a wide temperature range and broadened $\Delta T_{\frac{1}{2}}$. The optical inhomogeneity of the crystal we used also contributes to the broadened width of $\Delta T_{\frac{1}{2}}$. Under polarized light microscope, the interference pattern changes somewhat as the crystal is moved. This indicates that, at different locations on the crystal, there may actually be different phase matching temperatures.

Finally, we have measured the frequency doubling conversion efficiency. The conversion efficiency of the first frequency doubling is approximately 5% with the approximate energy of the 5300 \AA laser light being 100 mJ and the peak power being $\sim 2\text{ MW}$. After the loss due to absorption, reflection and scattering by the inserted filters and other optical components, the actual energy of the 5300 \AA light incident on the ADP crystal is only several tens of millijoules. Still using the quartz prism in the spectrograph for light diffraction, the 2650 \AA ultraviolet laser light energy is measured with an energy meter produced by the Chinese Measurement Science Research Institute accompanied by a sensitive detector. The ultraviolet laser energy was found to be 0.2 mJ. Due to the high sensitivity of the measuring instrument, small variation in ambient temperature causes large drift of the zero point. The experimental error is large and the result is only meaningful in its order of magnitude. It should also be pointed out that the energy meter is not calibrated for laser spectrum with wavelength less than 3000 \AA .

and the loss due to diffraction has not been carefully measured. We can therefore only estimate the order of magnitude of the generated ultraviolet laser energy to be 0.1 mJ and the conversion efficiency of the second frequency doubling roughly 1%. The results reported here are only preliminary and the experiment can be improved substantially. The 5% conversion efficiency of the first doubling of the frequency is below our best result. Generally, a 10% efficiency can be achieved. The second frequency doubling can also be made much more efficient by improving various stages of the experiment.

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